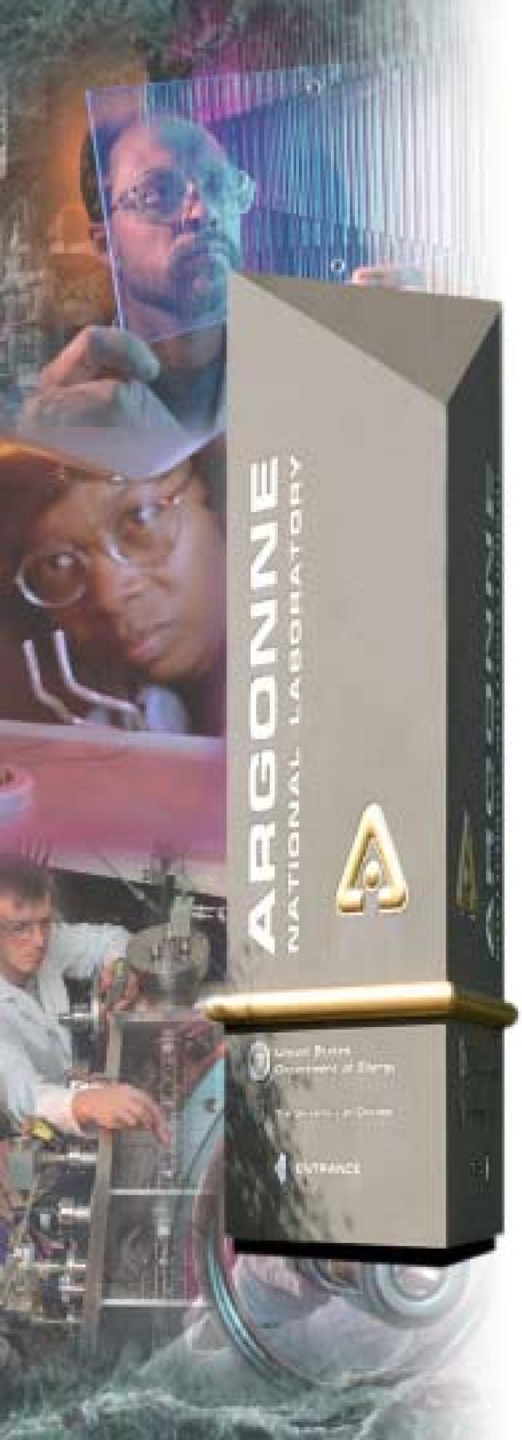


Time-resolved X-ray Diffraction Imaging of Ferroelectric Domains in $BaTiO_3$ Single Crystals

Yong S. Chu and Andrei Tkachuk

Experimental Facilities Division, Argonne National Laboratory, IL

Workshop on
Emerging Scientific Opportunities Using X-ray Imaging

Argonne National Laboratory

Office of Science
U.S. Department of Energy

A U.S. Department of Energy
Office of Science Laboratory
Operated by The University of Chicago

Two Other Talks TODY on this Subject

12:00 “X-ray Studies of Ferroelectrics”
Carol Thompson
Workshop on Mesoscopic and Manoscopic Science

16:30 “Nanosecond Switching Dynamics in Ferroelectric Devices”
Paul Evans
Workshop on Time-Domain Science

Acknowledgement

- Francesco De Carlo (XOR, ANL),   Full-Field Program at 2-BM

- Ferroelectric Domains in BTO:
  Hoydoo You and Andreas Menzel (MSD, ANL)
  Martin Holt (8-ID-E/McGill University)

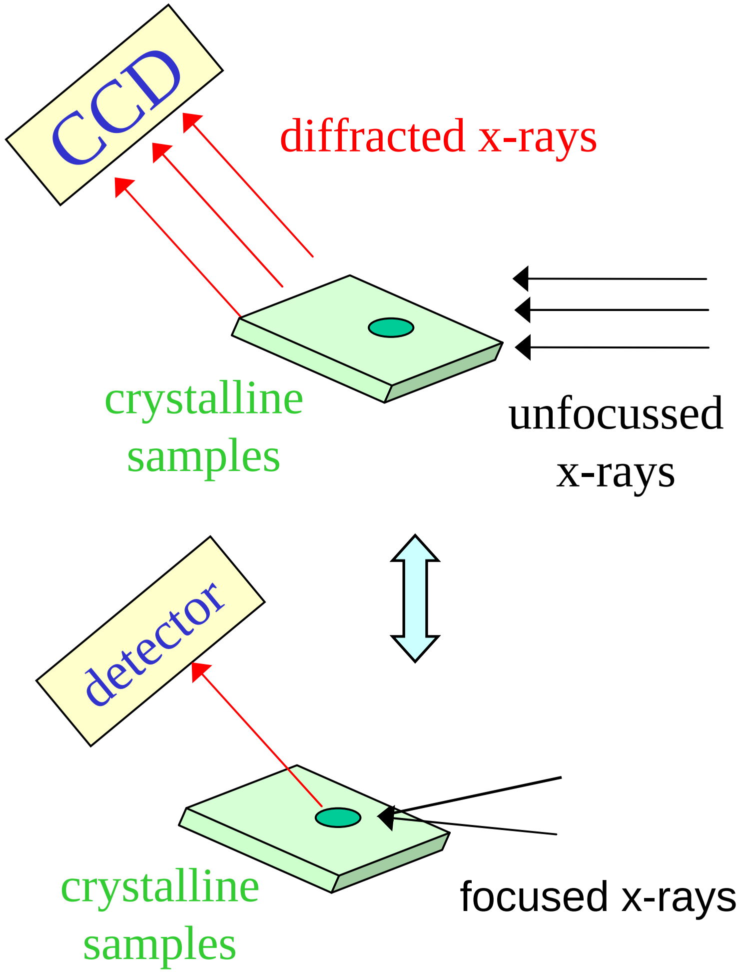
- In-Situ Tomography and Diffraction
  Francesco De Carlo
  Ersan Üstündag (Iowa State University)
  Jay Hanan **(**Oklahoma State University)

- Investigation of Defects in Protein Crystals:
  Zhengwei Hu (NASA/Marshall Space Flight Center), Barry Lai (XOR, ANL)

- Diamond Etching
  Albert Macrander, Felix KRASNICKI, Yuncheng Zhong, XFD, ANL

- Damage to Si induced by Femto-Second Laser
  Andy Winholtz (U. of Missouri)

X-ray Topography? Diffraction Imaging?



Diffraction Microscopy

- resolution
  spatial vs. angular
- sensitivity
- throughput
- contrast "enhancement" with coherent x-rays

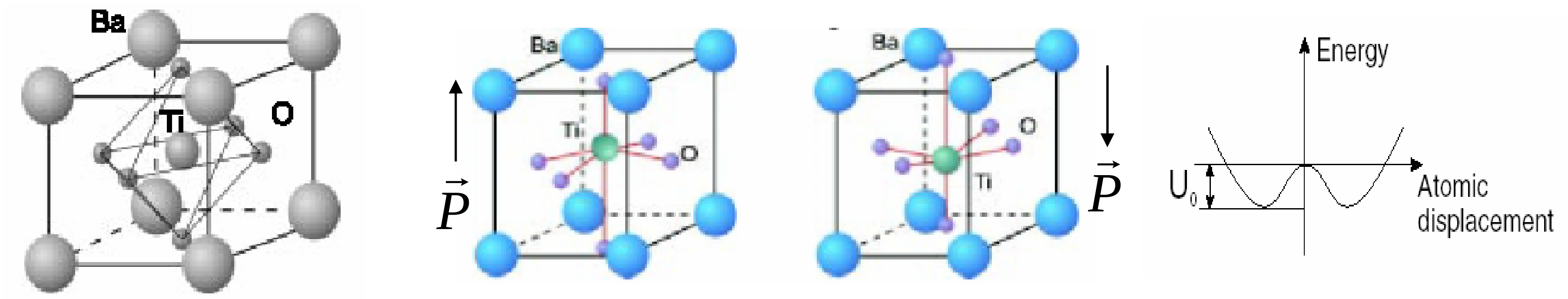
Outline

- Background on ferroelectric domains
- Experimental Results
- Future

Background: Ferroelectric Materials

• Perovskites ($BaTiO_3$, $PbTiO_3$, $LiNbO_3$ …), KDP type (KH_2PO_4, …), TGS type (Tir-glycine sulfate, …)

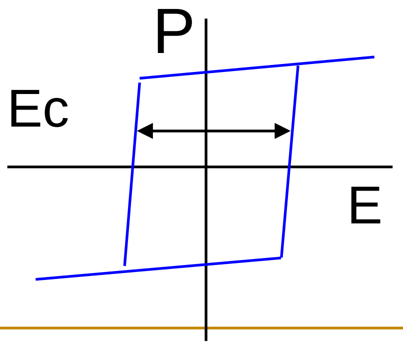
• Spontaneous polarization due to the loss of centro-symmetry below Tc



cubic at T > Tc

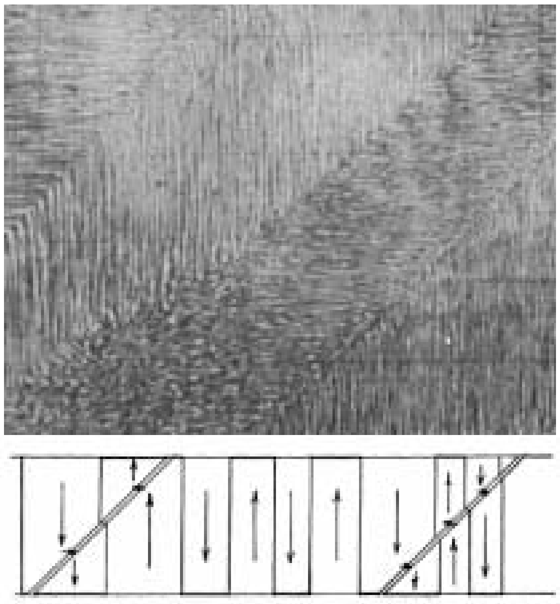
tetragonal at T < Tc

• Switchable electric polarization



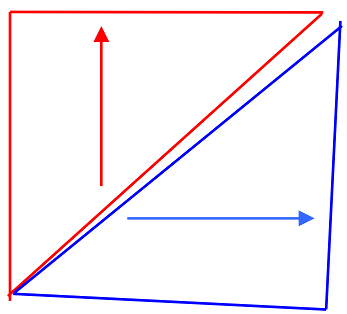
Background: Ferroelectric Domains

- FE domains form to lower electrostatic energy
- 90° and 180° domains
- early investigations involved BTO single crystals
- recent investigations focused on thin-films

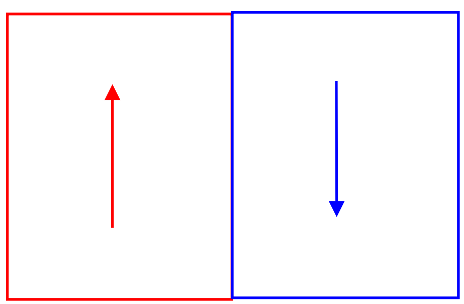


Hooton and Merz, Phys. Rev, 98 (1955)

c>a

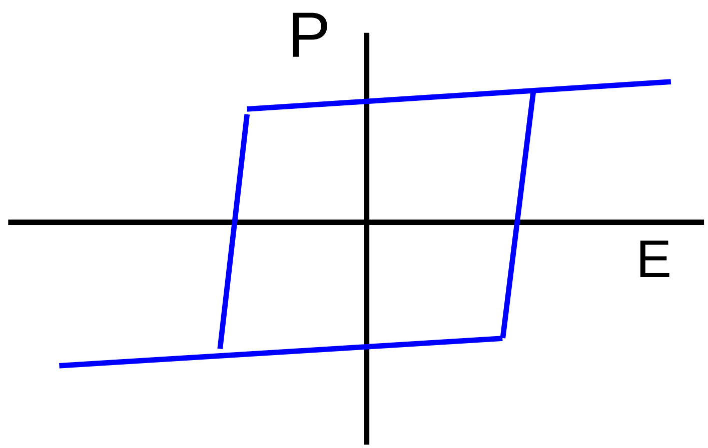


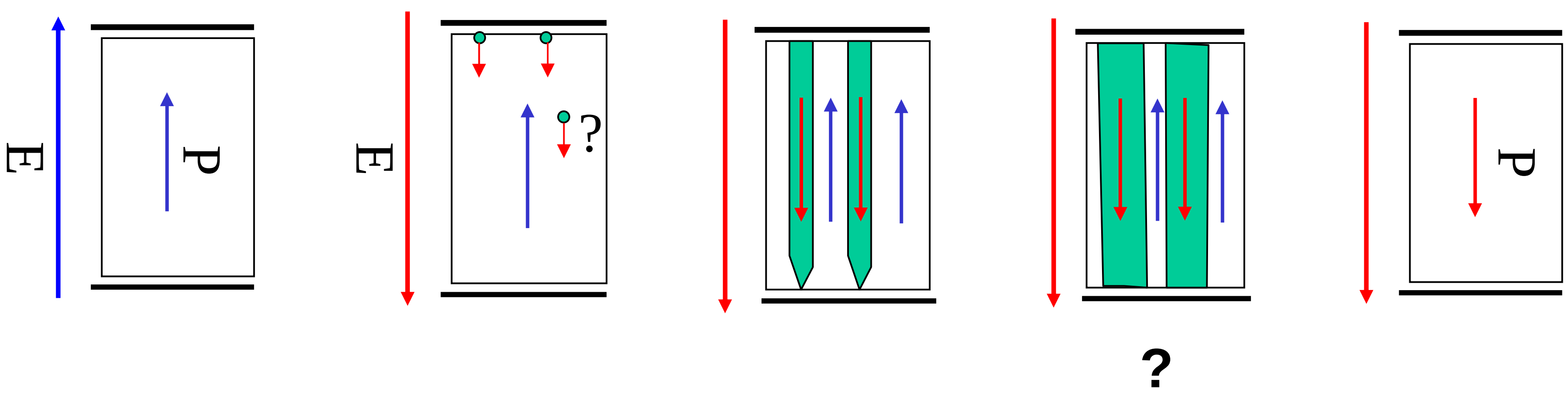
90° domains



180° domains

Background: Generally Accepted Polarization Reversal Process





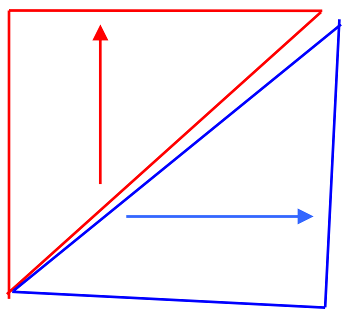
Background: Unresolved Problems

- 180° domain wall thickness:
  - early calculations: a few lattice parameters
  - TEM: 10~100 A
  - theoretical consideration of defects: 300~3000A.
  - x-ray topography: 1~5 μm → nonferroeleastic walls due to strain?
  - increased visibility of moving wall

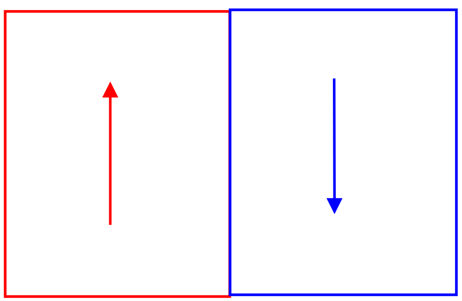
- nucleation and growth
  - theoretic values for critical nucleation energy is too high.
    → defects play predominant role.
  - virtually no data on nucleation and growth for E < Ec
  - dislocation/stress during growth.

Background: Diffraction Contrast of Domains

c/a ~1.01



90° domains



180° domains

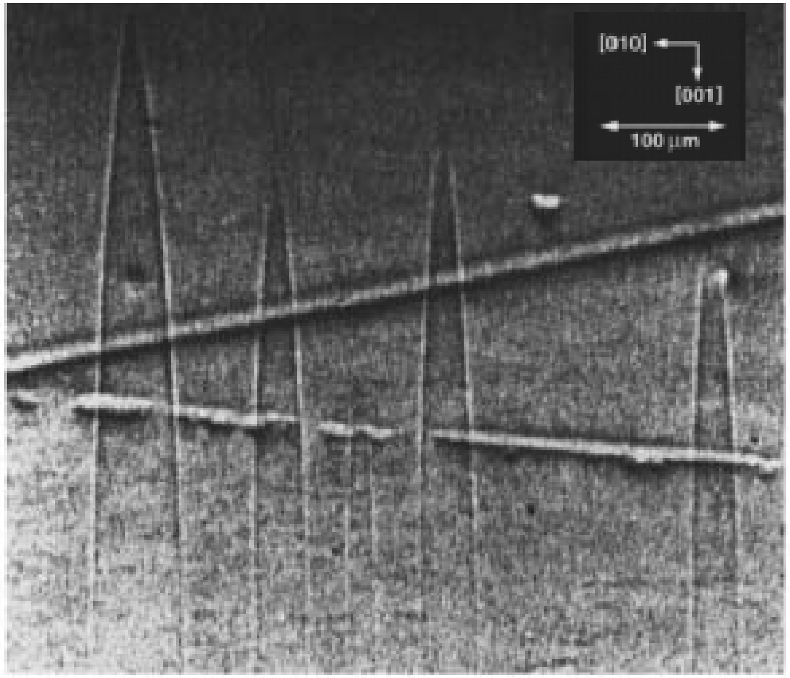
orientation & d-spacing

same orientation and d-spacing

loss of centro symmetry

$F_{hkl} \neq F_{-h-k-l}$

Background: Diffraction Contrast of 180° Domain Walls

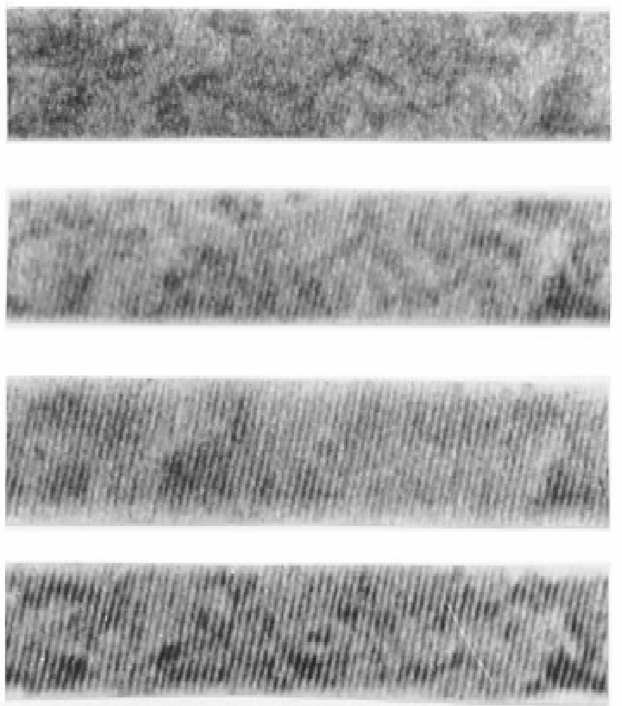


BTO

- **Sheer-Strain Field** at the domain wall:
  Kobayashi,et.al. PRL, 11 (1963)
  Kawata. et. al. J. Phys. Soc. Jap, 50, 3398 (1981).
- **Dielectric Constant Difference** (pure dynamical effect)
  Forgarty et. Al. J. Opt. Soc. Am., 13, 2636 (1996).

Background: Diffraction Contrast of 180° Domain Walls

LibNbO$_3$

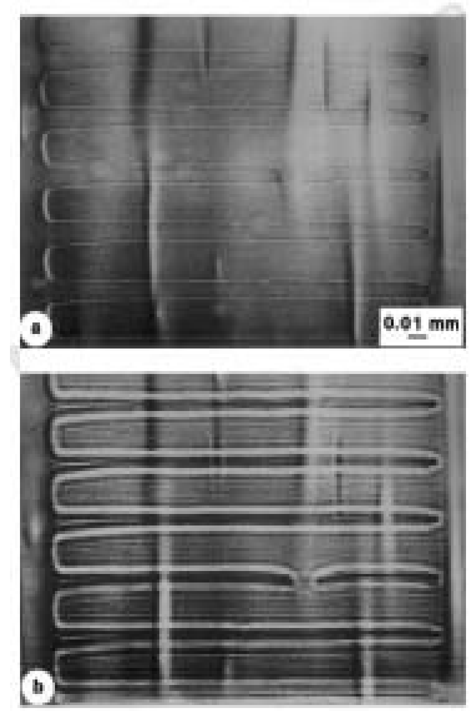


D=0.11 m

D=0.51 m

D=1.11 m

D=1.67 m



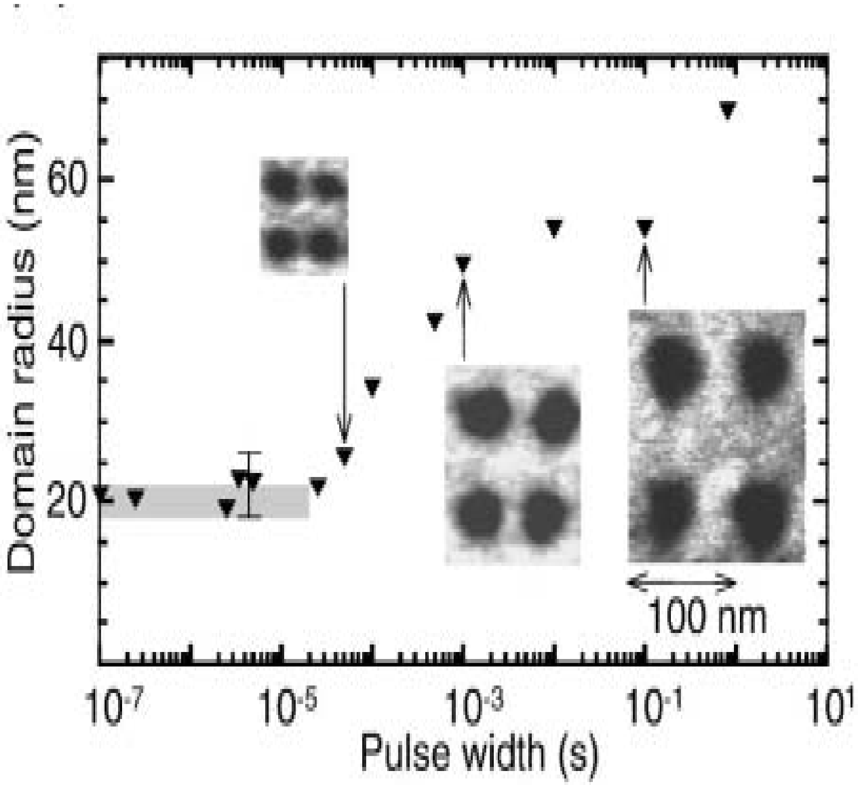
D=0.005 m

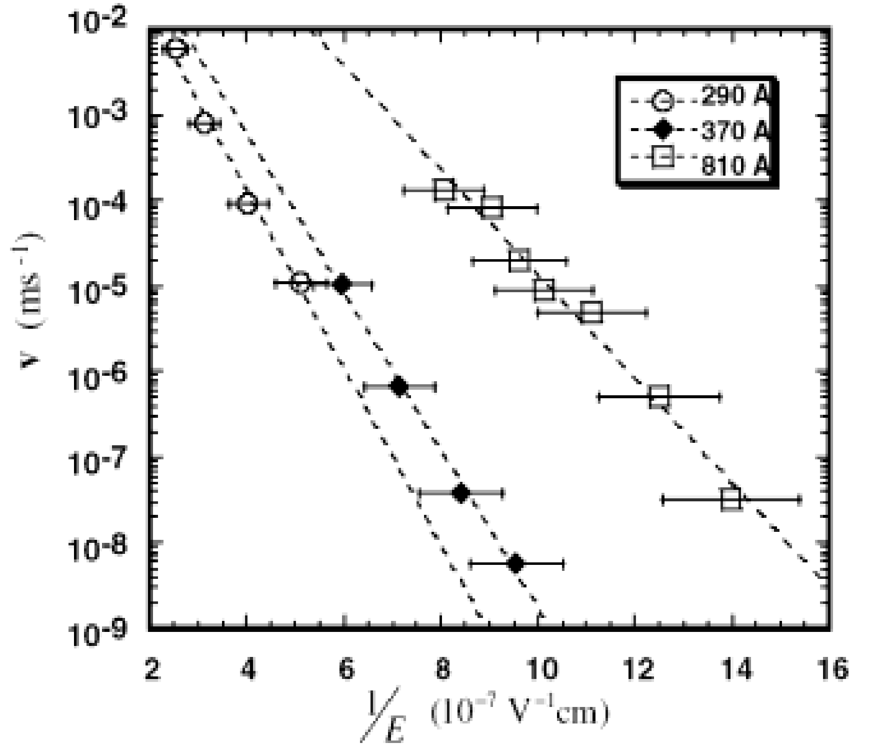
D=0.2m

phase-contrast due to
Phase-Shift between F_{hkl} and F_{-h-k-l}
Rejmankova-Pernot, P. Cloetens, J. Baruchel and J.-P. Guigay, PRL, 81, 3435(1998)

Lattice Distortion "enhanced" by Phase-Contrast
Hu, et. al. Nature, 392, 690 (1998)

AFM written domains: $Pb(Zr_{0.2}Ti_{0.8})O_3/SrTiO_3(001)$

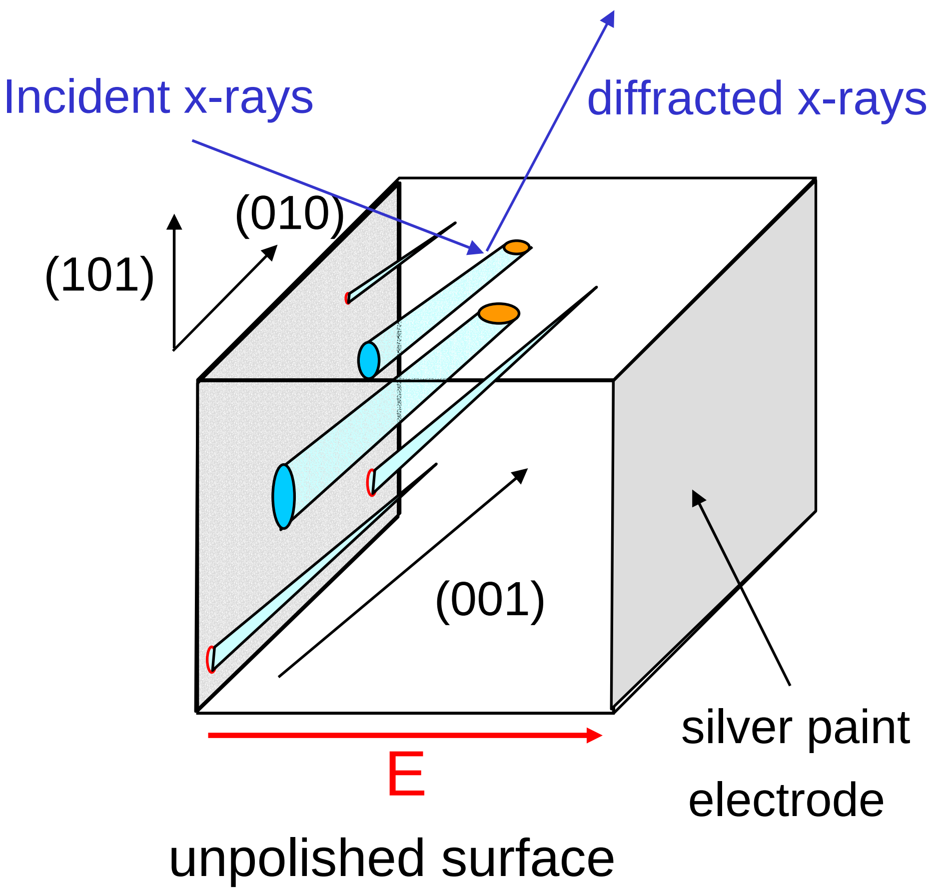




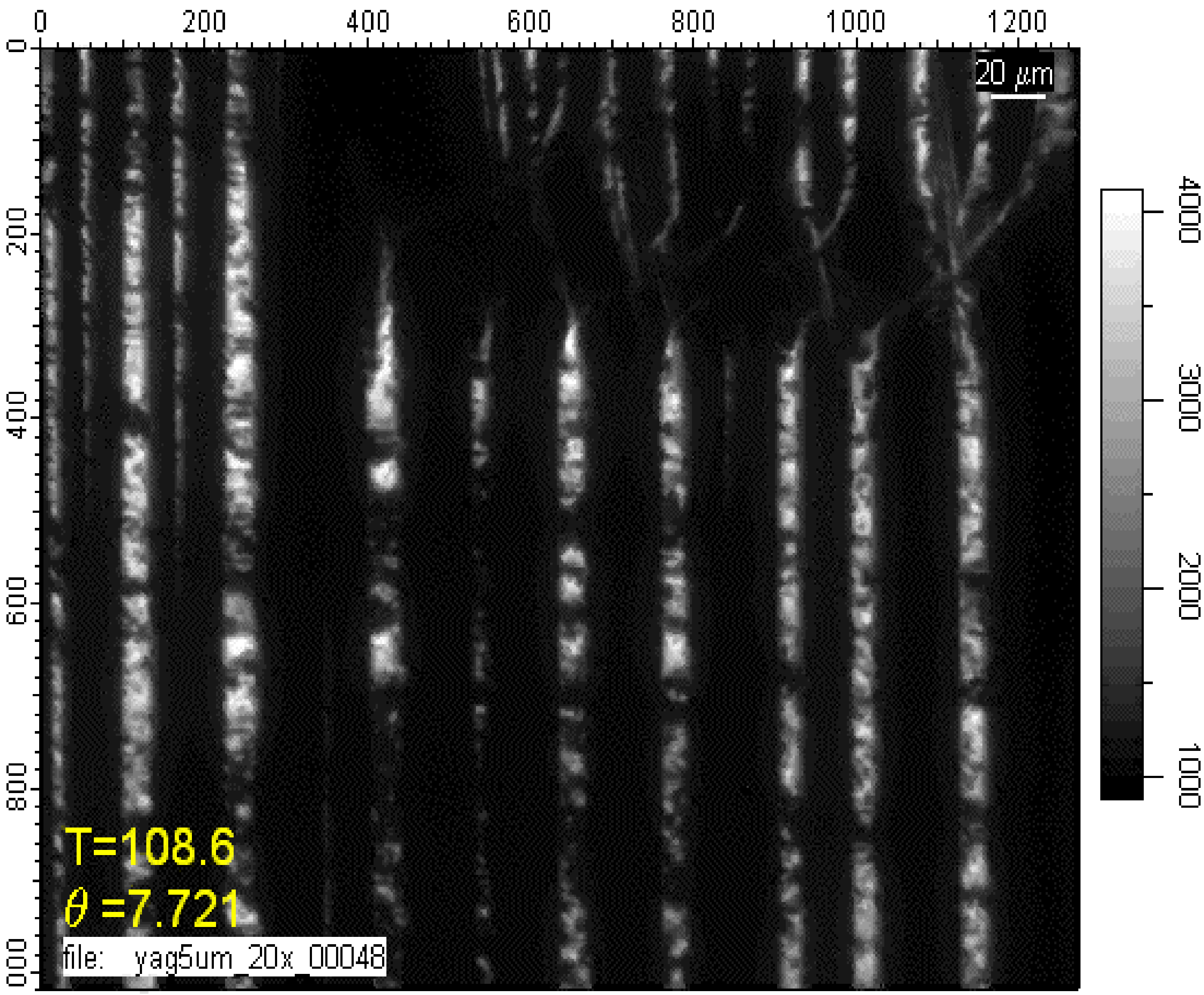
Tybell et. Al. PRL 89, 097601 (2002)

Two Experiments

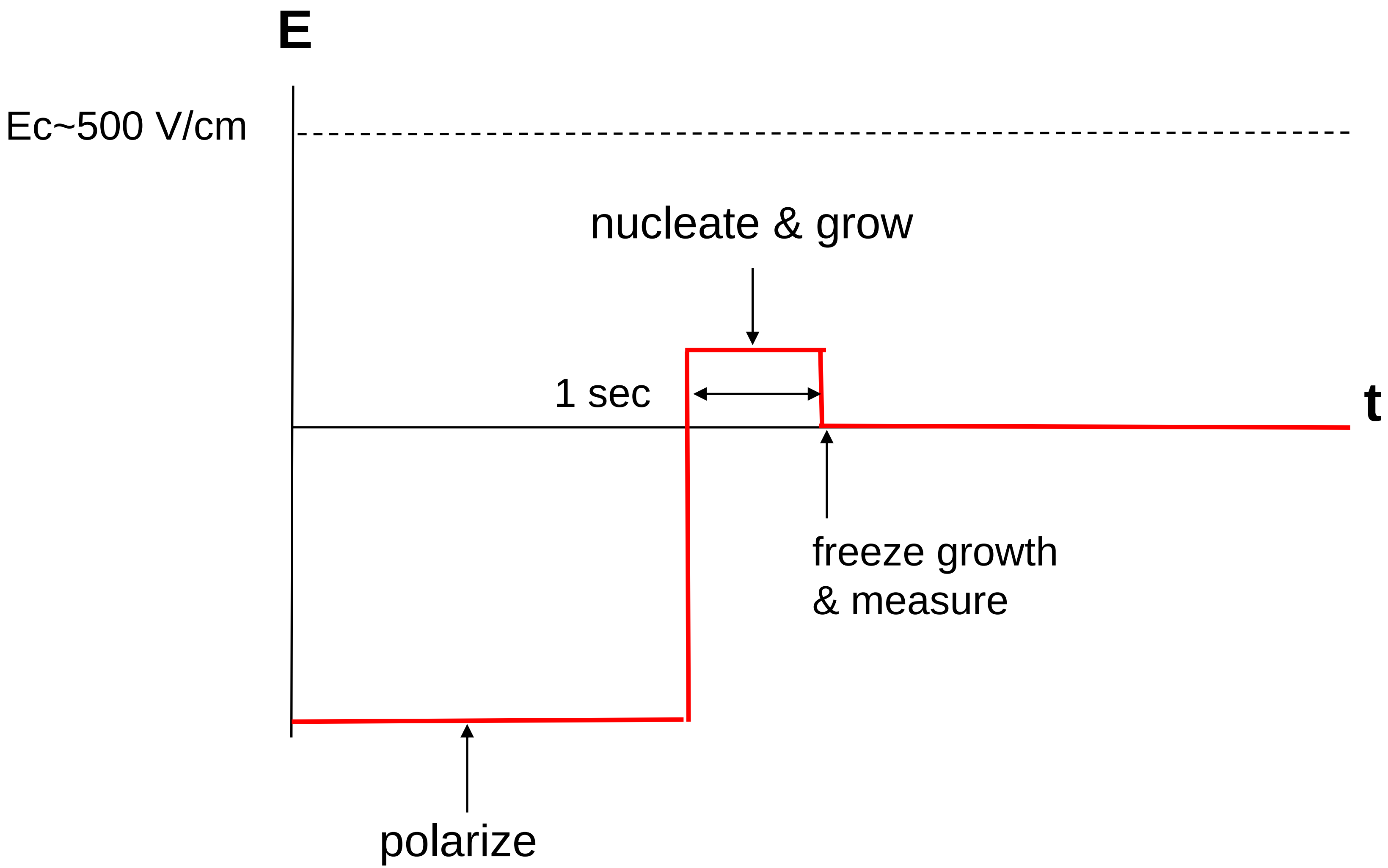
Experiment #1



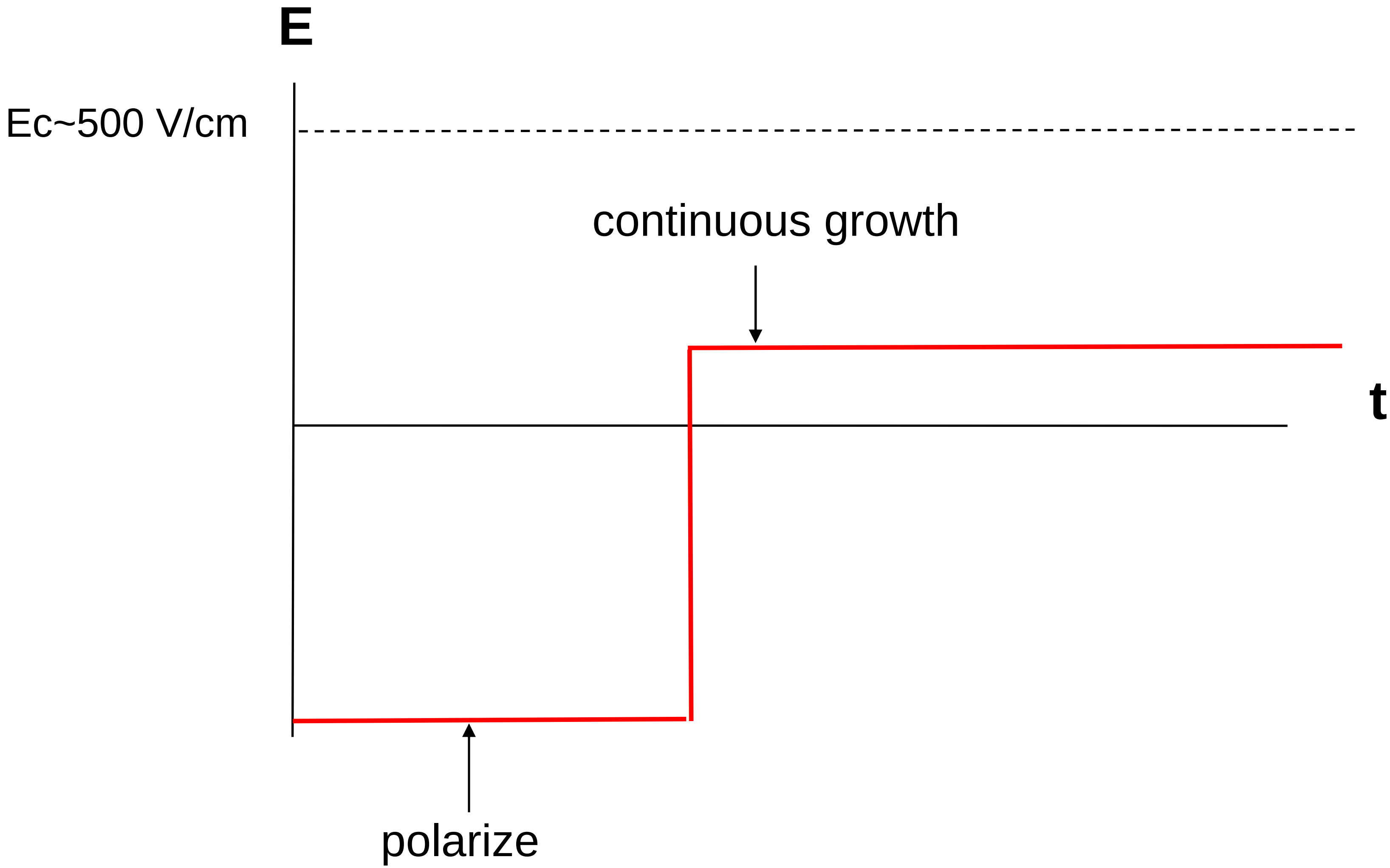
Nucleation of 90° Ferroelectric Domians
Potential Ramped Down from 2 keV/cm to 0



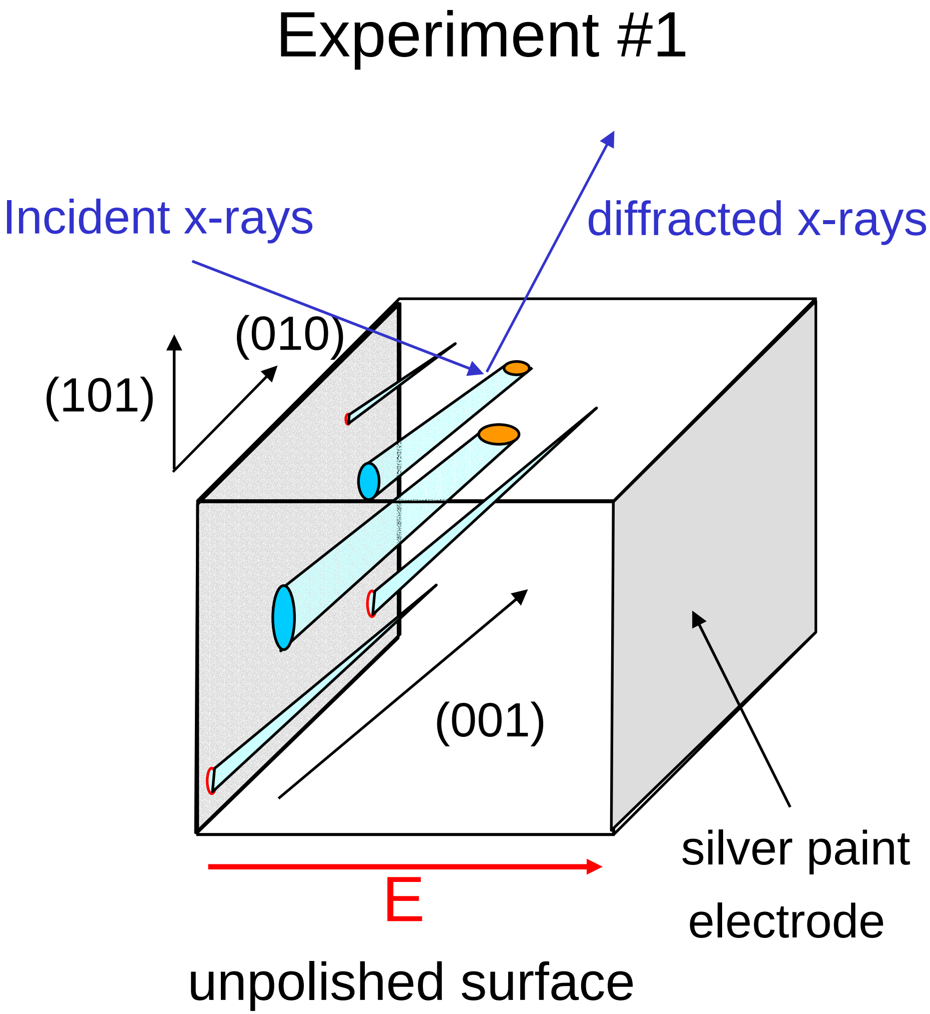
Measurement of Domain Growth

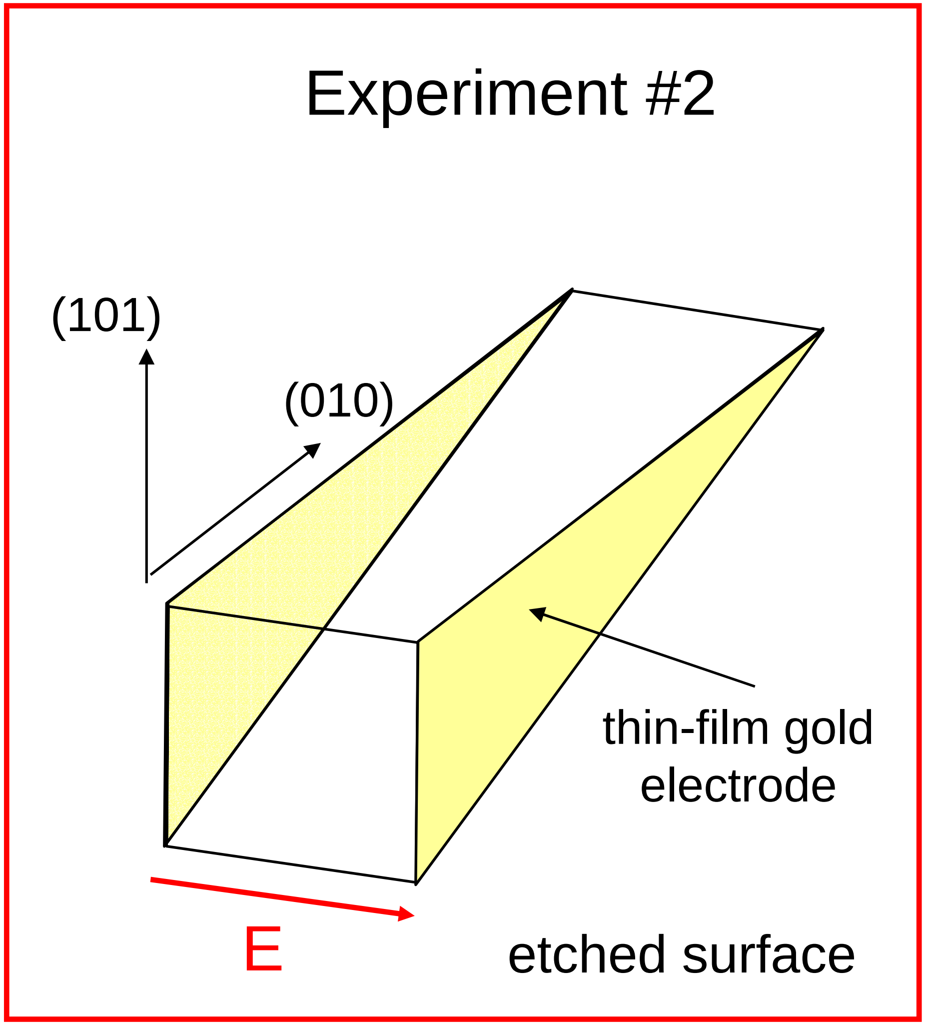


Measurement of Domain Size vs. Time

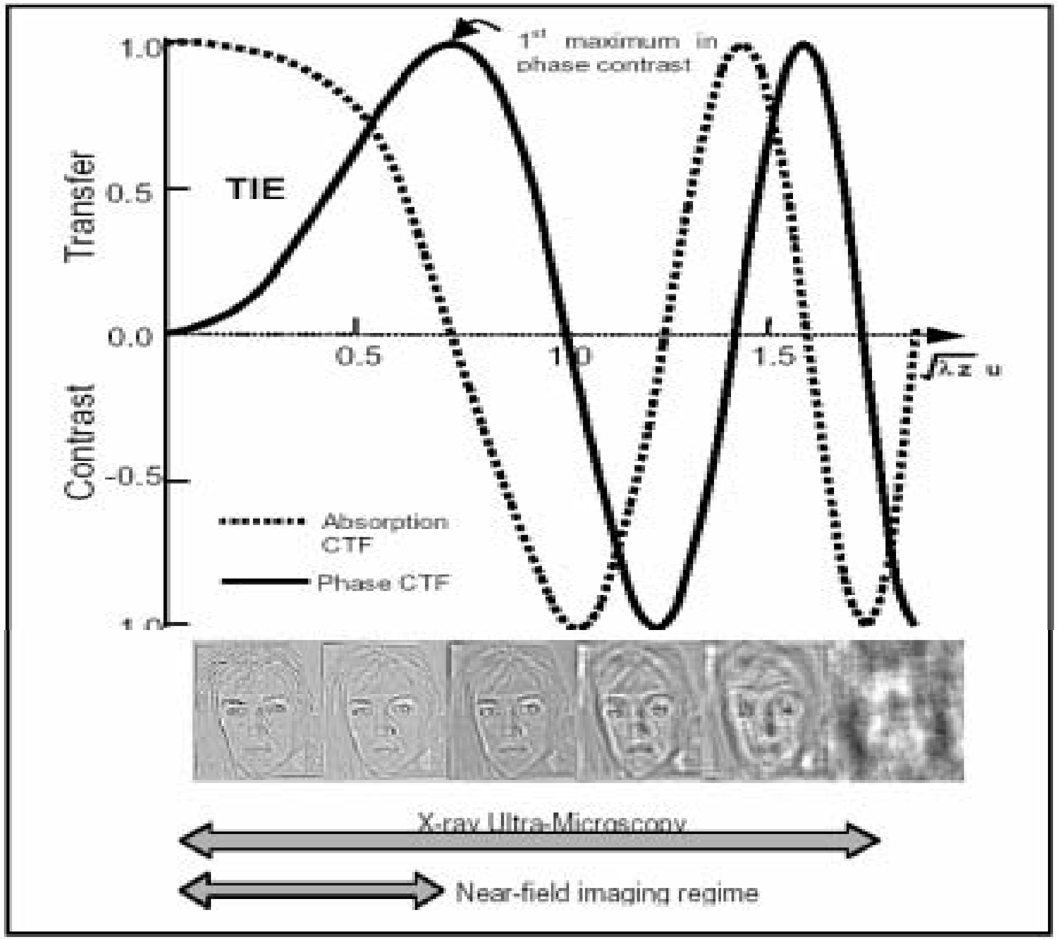


Two Experiments





Enhancement of Visibility: Phase-Contrast Function

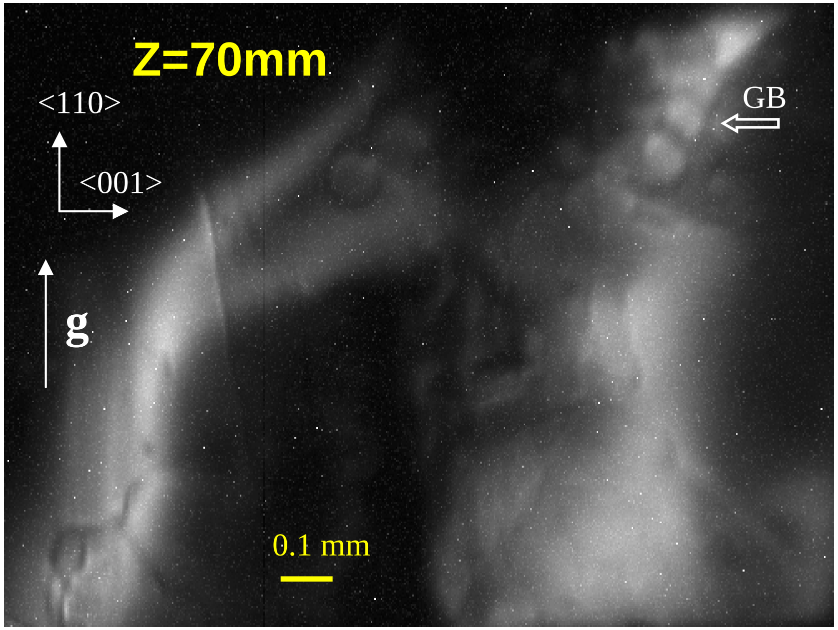


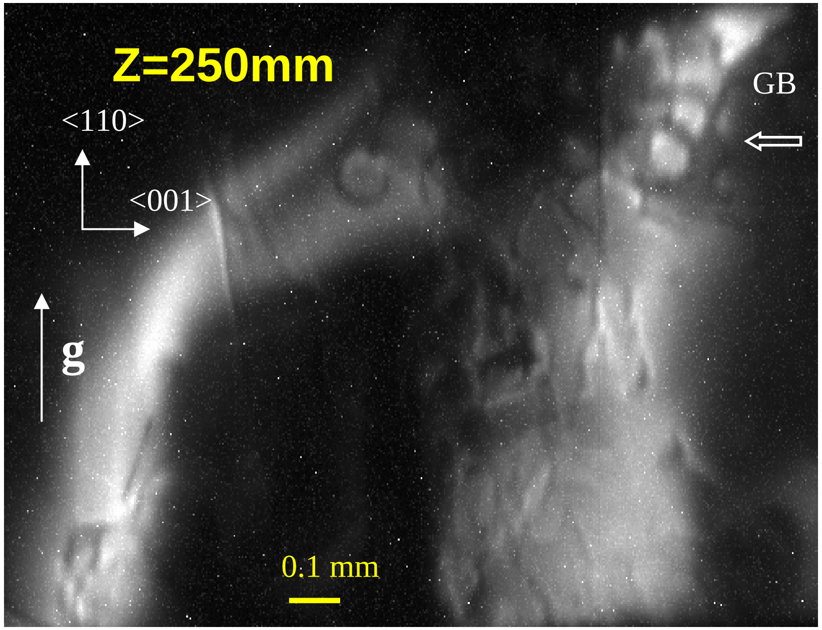
Mayo et. al. Optics Express 11, 2289 (2003)

$$z = D^2 / \lambda$$

Phase-Contrast "Bragg" Diffraction Imaging

Lysozime Protein Crystal





PRL 87, 148101 (2001)

What's Next

1) Further investigate the origin of the diffraction contrast at the domain wall.

2) Measure electrode/sample interface.

3) Find way to do it FASTER.

Challenges

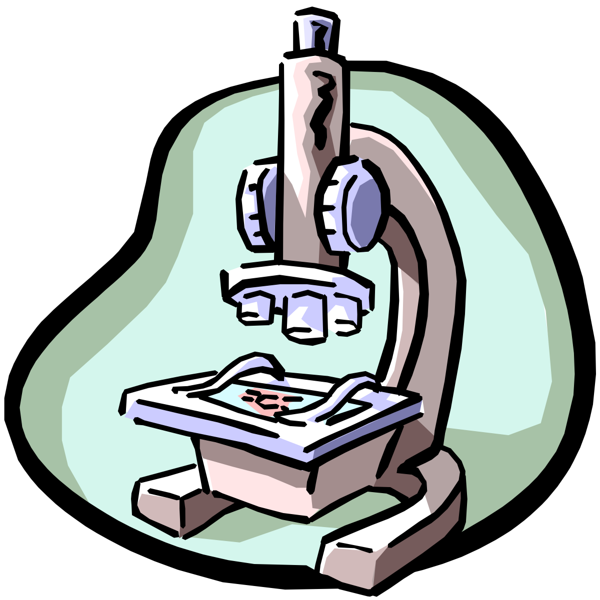
1) Quantitative Phase Retrieval from Diffraction Imaging

Can one quantify,

strain, refraction, and pure phase-contrast?

Challenges

2) Integrated Approach for X-ray Microscopy



- **Full-Field vs. Scanning**
  - field of view
  - throughput
  - sensitivity
  - resolution
- **In-situ Application**
  - “combo” approach

How can we utilize the BEST capabilities from Both Methods?